



## INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

<b>(51) International Patent Classification <sup>5</sup> :</b> <b>C10K 3/00, C01B 3/24</b> <b>C09C 1/48</b>	<b>A1</b>	<b>(11) International Publication Number:</b> <b>WO 93/12205</b> <b>(43) International Publication Date:</b> 24 June 1993 (24.06.93)
<b>(21) International Application Number:</b> PCT/NO92/00200 <b>(22) International Filing Date:</b> 11 December 1992 (11.12.92) <b>(30) Priority data:</b> 914909 12 December 1991 (12.12.91) NO <b>(71) Applicant (for all designated States except US):</b> KVÆRNER ENGINEERING A.S [NO/NO]; Prof. Kohtsvei 5, N- 1324 Lysaker (NO). <b>(72) Inventors; and</b> <b>(75) Inventors/Applicants (for US only) :</b> LYNUM, Steinar [NO/ NO]; Hartmannsvei 40 B, N-0284 Oslo (NO). HAUG- STEN, Kjell [NO/NO]; General Krohgs vei 8, N-0379 Oslo (NO). HOX, Ketil [NO/NO]; Gjørtlerlvn. 10, N- 7021 Trondheim (NO). HUGDAHL, Jan [NO/NO]; Ha- rald Botnersveg 10, N-7011 Trondheim (NO). MYKLE- BUST, Nils [NO/NO]; Nidarøyt. 6B, N-7030 Trond- heim (NO).		<b>(74) Agent:</b> ONSAGERS PATENTKONTOR AS; P.O. Box 265 Sentrum, N-0103 Oslo (NO).  <b>(81) Designated States:</b> AT, AU, BB, BG, BR, CA, CH, CS, DE, DK, ES, FI, GB, HU, JP, KP, KR, LK, LU, MG, MN, MW, NL, NO, NZ, PL, PT, RO, RU, SD, SE, UA, US, European patent (AT, BE, CH, DE, DK, ES, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE), OAPI patent (BF, BJ, CF, CG, CI, CM, GA, GN, ML, MR, SN, TD, TG).  <b>Published</b> <i>With international search report.</i>
<b>(54) Title:</b> A METHOD FOR COMBUSTION OF HYDROCARBONS  <b>(57) Abstract</b> <p>In order to enrich natural gas or other hydrocarbon gases with hydrogen, thus reducing the carbon content and thereby achieving a reduction or elimination of the discharge of carbon dioxide during combustion of the gases, a pyrolytic process is conducted in the feed stream for the natural gases or hydrocarbon gases which are to be burned. The gas is passed through a reactor in which it is decomposed at least partially into a carbon constituent and a hydrogen constituent. The carbon constituent is removed to a desired level. Any remaining constituent together with the hydrogen constituent is conveyed to the combustion process while the removed carbon constituent is conveyed out of the process for separate application. The pyrolytic process can be carried out on the entire stream or only on a partial stream.</p>		

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A method for combustion of hydrocarbons

The invention concerns a method for reducing the carbon content in a feed flow of natural gas or other hydrocarbon gases for a combustion process or a chemical process in order thereby to eliminate to the desired degree or at least to reduce the discharge of carbon dioxides in the combustion of the gas, e.g. in connection with a gas-fired power station.

During the combustion of fossil fuels, coal, natural gas and other hydrocarbons, both water in the form of vapour and carbon dioxide will be found in the waste gases. The proportional incidence between these two gases will be dependent on the quantitative ratio of carbon to hydrogen in the fuel concerned. Coal will produce almost only carbon dioxide, methane will produce water and carbon dioxide in the ratio of 2:1, while hydrogen will only produce water. In the efforts to reduce the greenhouse effect, the discharges of carbon dioxide are becoming ever less acceptable. Thus it is vitally important to limit these discharges, the best best solution being to eliminate them entirely.

The object of the present invention is to improve this situation by providing a method for reducing the carbon content of natural gas and hydrocarbon gas. This reduction should be capable of being implemented in such a manner that a desired degree of reduction of the CO<sub>2</sub> content can be achieved after a combustion or a chemical process. The reduction can extend all the way through to the use of pure hydrogen as a fuel, thus completely avoiding the discharges of CO<sub>2</sub>.

Thus a further object of the invention is also to make it possible for natural gas to be used as a fuel with a greatly reduced production of carbon dioxide, e.g. in a gas turbine.

These objects are achieved by a method which according to the invention is characterized by the features in the claims presented.

The invention is based on the fact that it is possible to decompose hydrocarbons pyrolytically into carbon and hydrogen. By using pure hydrogen as a fuel the discharge of carbon dioxide will be eliminated. If carbon is removed from the natural gas or the hydrocarbon gas which is used as a fuel, the discharge of carbon dioxide will be reduced before the combustion.

A reduction of this kind can be carried out by performing a complete or partial conversion of natural gas or hydrocarbon gases in a feed stream to a combustion process or a chemical process. The feed stream is decomposed to the desired degree into carbon and hydrogen, whereof the natural gas with the reduced carbon content goes to combustion or a chemical process and the carbon constituent is removed from the process for separate application. Such a reduction can also be performed by converting a partial stream of the natural gas or the hydrocarbon gas.

In both cases the decomposition into carbon and hydrogen will require energy. The energy content in the gases with reduced carbon content is lower than in pure natural gas or hydrocarbon gas since the liberated carbon represents the lost energy. The result will be that more gas in total has to be supplied to the process in order to achieve the same net effect as from pure natural gas or hydrocarbon gas. The extent of this additional amount will be dependent on the degree to which the carbon content has to be reduced, i.e. the extent to which a conversion has to be performed, and also on the efficiency of the combustion process concerned.

The object is to be able to use pure hydrogen as a combustion gas in order thereby to be able to completely eliminate the CO<sub>2</sub> discharges.

At existing plants, however, there will be operating conditions

which prevent the use of pure hydrogen, thus making it necessary to add a certain amount of hydrocarbons.

The process is therefore designed so as to offer the possibility of adding a certain amount of natural gas or hydrocarbon gas to the hydrogen stream and the amount can be adjusted according to the operating conditions.

After a combustion process, the waste gases are purified by methods known in the art and the purification methods are often extremely expensive, as for example in the case of catalytic purification.

It has now been discovered that the expenses involved in the conversion of natural gas or hydrocarbon gas which are used as a fuel for a combustion plant, i.e. before combustion, can be compensated for, thus enabling an economically justifiable process to be achieved despite the energy loss in the combustion stream. This is achieved by producing pure carbon in addition to hydrogen in the pyrolytic process in the method according to the invention. The carbonaceous material will be present in the form of carbon black and as such a material will have a very high value. Carbon black can, e.g., be used as a reduction agent in the metallurgical industry or on the conventional "carbon black market". If the carbonaceous material is used for anodes in the aluminium industry, this will entail substantial improvements both from the environmental and the economic point of view, since the new anodes will not pollute either the electrolyte and thus the product or the environment by the discharge of sulphur and tar.

Thus by means of the invention a highly favourable combination has been obtained of financially profitable industrial production and conservation of the environment, i.e. a reduction in pollution.

In the following section the invention will be described in more detail by means of an example of the use of the method.

As an example, the operation of a gas-fired power station based on natural gas as its energy source has been chosen. The example is intended to illustrate the main principles of the invention. Other applications of the invention could be the production of pure hydrogen and the use of the method in all processes where natural gas or another hydrocarbon are used as the energy source. The invention is particularly well suited in connection with power production with fuel cells based on hydrogen as the means of combustion.

In the supply line to a gas-fired power station, which, e.g., is operated by the use of natural gas, e.g. methane, there is provided a reactor which is operated according to the pyrolytic principle with a plasma torch and which causes a decomposition of the incoming natural gas or hydrocarbon gas into a constituent of hydrogen and a constituent of carbon particles. The reactor may be located in a branch line or a line bypassing the reactor. The method of operation of such a reactor and the design of a plasma torch for this are described in the applicant's simultaneous Norwegian patent applications 91 4904 and 91 4907. As described in these documents, a quality control can be performed for the carbon constituent. The hydrogen formed is also used via a return line as a plasma-forming gas for the torch, thus enabling this entire reactor to be operated without causing any pollution.

The hydrogen gas from the reactor is then passed to the gas turbine where it is used either alone or together with hydrocarbons (methane) as fuel in the gas turbine. The gas turbine is operated in a conventional manner. However, there is one major difference, which is that the discharge of carbon dioxide is reduced or completely eliminated. The degree to which the discharges of carbon dioxide are reduced will be dependent on how large a proportion of the supply to the gas turbine is composed of pure hydrogen. Throughout the process,

the proportion can be regulated in the entire area from no admixture of hydrogen to pure hydrogen.

**PATENT CLAIMS**

1. A method for reducing the carbon content in a feed stream of natural gas or other hydrocarbon gases for a combustion process or a chemical process in order thereby to achieve a reduction in or elimination of the discharge of carbon dioxide during combustion or other chemical application of the gas, characterized in that the natural gas or hydrocarbon gas which is to undergo combustion is completely or partially subjected to a pyrolytic process, all gas or a part of the feed gas being passed through a reactor wherein it is completely or partially decomposed to the desired degree into a hydrogen constituent and a carbon constituent, that the carbon constituent together with any undecomposed constituent from the reactor and any separate constituent of the natural gas or the hydrocarbon gas is conveyed to a combustion process or a chemical process, and that the precipitated carbon constituent is conveyed out of the process for separate application.
2. A method according to claim 1, characterized in that parts of the hydrogen formed are recycled for use in the reactor during the pyrolytic process.
3. A method according to claim 1, characterized in that the feed stream is divided into two partial streams and that only one partial stream is passed through the reactor.
4. A method according to claim 1, characterized in that the carbon constituent which is removed is composed of pure carbon.
5. Application of a method as described in claim 1 for the extraction of pure hydrogen for use, e.g., in fuel cells or as fuel in internal combustion engines.

6. Application of the method according to claim 1 in connection with gas-fired power stations.
7. Application of the method according to claim 1 in connection with the combustion of hydrocarbons for energy purposes.
8. Application according to claim 3 for the extraction of carbon for the production of pure carbon for use, e.g. as a reduction material in the metallurgical industry.

## INTERNATIONAL SEARCH REPORT

International application No.

PCT/NO 92/00200

## A. CLASSIFICATION OF SUBJECT MATTER

IPC5: C10K 3/00, C01B 3/24, C09C 1/48

According to International Patent Classification (IPC) or to both national classification and IPC

## B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

IPC5: C10K, C01B, C09C

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

SE,DK,FI,NO classes as above

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)

## C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	DD, A, 211457 (HOFFMANN, HEINZ ET AL), 11 July 1984 (11.07.84), page 3, line 32 - page 5, line 9	1-4
X	DD, A1, 276098 (VEB CHEMIEANLAGENBAUKOMBINAT LEIPZIG-GRIMMA-STAMMBETRIEB), 14 February 1990 (14.02.90), page 2, line 52 - page 3, line 38	1-4

☒ Further documents are listed in the continuation of Box C.☒ See patent family annex.

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## C (Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	DE, A, 2413558 (SKF KUGELLAGERFABRIKEN GMBH), 17 October 1974 (17.10.74), page 2, line 19 - page 3, line 26  -----  -----	1-8

**INTERNATIONAL SEARCH REPORT**  
Information on patent family members

29/01/93

International application No.  
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Patent document cited in search report		Publication date	Patent family member(s)		Publication date
DD-A-	211457	11/07/84	NONE		
DD-A1-	276098	14/02/90	NONE		
DE-A-	2413558	17/10/74	AU-B-	475113	12/08/76
			AU-A-	6693074	25/09/75
			BE-A-	885257	16/01/81
			CA-A-	1007050	22/03/77
			FR-A,B-	2223449	25/10/74
			JP-C-	1065669	30/09/81
			JP-A-	50026794	19/03/75
			JP-B-	56004482	30/01/81
			SE-B,C-	371453	18/11/74